LETTERS

Magnetic monopoles in spin ice

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Electrically charged particles, such as the electron, are ubiquitous. In contrast, no elementary particles with a net magnetic charge have ever been observed, despite intensive and prolonged searches (see ref. 1 for example). We pursue an alternative strategy, namely that of realizing them not as elementary but rather as emergent particles—that is, as manifestations of the correlations present in a strongly interacting many-body system. The most prominent examples of emergent quasiparticles are the ones with fractional electric charge *e*/3 in quantum Hall physics[2]. Here we propose that magnetic monopoles emerge in a class of exotic magnets known collectively as spin ice[3–5]: the dipole moment of the underlying electronic degrees of freedom fractionalises into monopoles. This would account for a mysterious phase transition observed experimentally in spin ice in a magnetic field[6,7], which is a liquid–gas transition of the magnetic monopoles. These monopoles can also be detected by other means, for example, in an experiment modelled after the Stanford magnetic monopole search[8].

Spin-ice materials are characterized by the presence of magnetic moments $\boldsymbol{\mu}_i$ residing on the sites i of a pyrochlore lattice (depicted in Fig. 1). These moments are constrained to point along their respective local Ising axes $\hat{e}_i$ (the diamond lattice bonds in Fig. 1), and they can be modelled as Ising spins $\boldsymbol{\mu}_i = \mu S_i \hat{\mathbf{e}}_i$, where $S_i = \pm 1$ and $\mu = |\boldsymbol{\mu}_i|$. For the spin-ice compounds discussed here, $Dy_2Ti_2O_7$ and $Ho_2Ti_2O_7$, (where Dy is dysprosium and Ho is holmium) the magnitude μ of the magnetic moments equals approximately ten Bohr magnetons ($\mu \approx 10\mu_B$). The thermodynamic properties of these compounds are known to be described with good accuracy by an energy term that accounts for the nearest-neighbour exchange and the long-range dipolar interactions[9,10] (for a review of spin ice, see ref. 4):

$$H = \frac{J}{3}\sum_{\langle ij \rangle} S_i S_j + Da^3 \sum_{(ij)} \left[\frac{\hat{e}_i \cdot \hat{e}_j}{|\mathbf{r}_{ij}|^3} - \frac{3(\hat{e}_i \cdot \mathbf{r}_{ij})(\hat{e}_j \cdot \mathbf{r}_{ij})}{|\mathbf{r}_{ij}|^5} \right] S_i S_j \qquad (1)$$

The distance between spins is r_{ij}, and $a \approx 3.54$ Å is the pyrochlore nearest-neighbour distance. $D = \mu_0 \mu^2/(4\pi a^3) = 1.41$ K is the coupling constant of the dipolar interaction.

Spin ice was identified as a very unusual magnet when it was noted that it does not order to the lowest temperatures T even though it appeared to have ferromagnetic interactions[3]. Indeed, spin ice was found to have a residual entropy at low T (ref. 5), which is well-approximated by the Pauling entropy for water ice, $S \approx S_P = (1/2)\log(3/2)$ per spin. Pauling's entropy measures the huge ground-state degeneracy arising from the so-called ice rules. In the context of spin ice, its observation implies a macroscopically degenerate ground state manifold obeying the 'ice rule' that two spins point into each vertex of the diamond lattice, and two out.

We contend that excitations above this ground-state manifold—that is, defects that locally violate the ice rule—are magnetic monopoles with the necessary long-distance properties. From the perspective of the seemingly local physics of the ice rule, the emergence of monopoles at first seems rather surprising. We will probe deeper into how the long-range magnetic interactions contained in equation (1) give rise to the ice rule in the first place. We then incorporate insights from recent progress in understanding the entropic physics of spin ice, and the physics of fractionalization in high dimensions[11–15], of which our monopoles will prove to be a classical instance.

We consider a modest deformation of equation (1), loosely inspired by Nagle's work[16] on the 'unit model' description of water ice: we replace the interaction energy of the magnetic dipoles living on pyrochlore sites with the interaction energy of dumbbells consisting of equal and opposite magnetic charges that live at the ends of the diamond bonds (see Fig. 2). The two ways of assigning charges on each diamond bond reproduce the two orientations of the original dipole. Demanding that the dipole moment of the spin be reproduced quantitatively fixes the value of the charge at $\pm\mu/a_d$, where the diamond lattice bond length $a_d = \sqrt{3/2}\,a$.

The energy of a configuration of dipoles is computed as the pairwise interaction energy of magnetic charges, given by the magnetic Coulomb law:

$$V(r_{\alpha\beta}) = \begin{cases} \frac{\mu_0}{4\pi}\frac{Q_\alpha Q_\beta}{r_{\alpha\beta}} & \alpha \neq \beta \\ \frac{1}{2} v_o Q_\alpha^2 & \alpha = \beta \end{cases} \qquad (2)$$

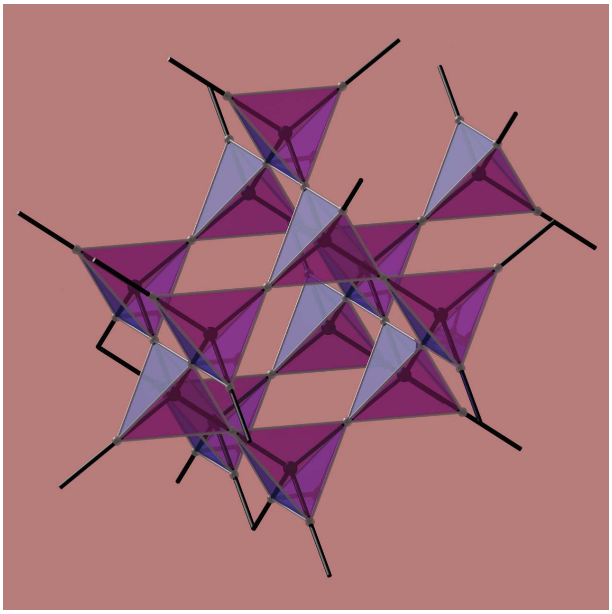

Figure 1 | The pyrochlore and diamond lattices. **The magnetic moments in spin ice reside on the sites of the pyrochlore lattice, which consists of corner-sharing tetrahedra. These are at the same time the midpoints of the bonds of the diamond lattice (black) formed by the centres of the tetrahedra. The ratio of the lattice constant of the diamond and pyrochlore lattices is $a_d/a = \sqrt{3/2}$. The Ising axes are the local [111] directions, which point along the respective diamond lattice bonds.**

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where Q_α denotes the total magnetic charge at site α in the diamond lattice, and $r_{\alpha\beta}$ is the distance between two sites. The finite 'self-energy' $v_0/2$ is required to reproduce the net nearest-neighbour interaction correctly. Equation (2)—which is derived in detail in the Supplementary Information—is equivalent to the dipolar energy equation (1), up to corrections that are small everywhere, and vanish with distance at least as fast as $1/r^5$.

We consider first the ground states of the system. The total energy is minimized if each diamond lattice site is net neutral, that is, we must orient the dumbbells so that $Q_\alpha = 0$ on each site. But this is just the above-mentioned ice rule, as illustrated in Fig. 2. Thus, one of the most remarkable features of spin ice follows directly from the dumbbell model: the measured low-T entropy agrees with the Pauling entropy (which follows from the short-distance ice rules), even though the dipolar interactions are long-range.

We now turn to the excited states. Naively, the most elementary excitation involves inverting a single dipole / dumbbell to generate a local net dipole moment 2μ. However, this is misleading in a crucial sense. The inverted dumbbell in fact corresponds to two adjacent sites with net magnetic charge $Q_\alpha = \pm q_m = \pm 2\mu/a_d$—a nearest-neighbour monopole–antimonopole pair. As shown in Fig. 2e, the monopoles can be separated from one another without further violations of local neutrality by flipping a chain of adjacent dumbbells. A pair of monopoles separated by a distance r experiences a Coulombic interaction, $-\mu_0 q_m^2/(4\pi r)$, mediated by monopolar magnetic fields, see Fig. 3.

This interaction is indeed magnetic, hence the presence of the vacuum permeability μ_0, and not $1/\varepsilon_0$, the inverse of the vacuum permittivity. It takes only a finite energy to separate the monopoles to infinity (that is, they are deconfined), and so they are the true elementary excitations of the system: the local dipolar excitation fractionalizes.

By taking the pictures from the dumbbell representation seriously, we may be thought somehow to be introducing monopoles where there were none to begin with. In general, it is of course well known that a string of dipoles arranged head to tail realizes a monopole–antimonopole pair at its ends[17]. However, to obtain deconfined monopoles, it is essential that the cost of creating such a string of dipoles remain bounded as its length grows, that is, the relevant string tension should vanish. This is evidently not true in a vacuum (such as that of the Universe) where the growth of the string can only come at the cost of creating additional dipoles. Magnetic materials, which come equipped with vacua (ground states) filled with magnetic dipoles, are more promising. However, even here a dipole string is not always a natural excitation, and when it is—for example, in an ordered ferromagnet – a string of inverted dipoles is accompanied by costly domain walls along its length (except, as usual, for one-dimensional systems[18]), causing the incipient monopoles to remain confined.

The unusual properties of spin ice arise from its exotic ground states. The ice rule can be viewed as requiring that two dipole strings enter and exit each site of the diamond lattice. In a typical spin-ice ground state, there is a 'soup' of such strings: many dipole strings of arbitrary size and shape can be identified that connect a given pair of sites. Inverting the dipoles along any one such string creates a monopole–antimonopole pair on the sites at its ends. The associated energy cost does not diverge with the length of the string, unlike in the case of an ordered ferromagnet, because no domain walls are created along the string, and the monopoles are thus deconfined.

We did not make reference to the Dirac condition[19] that the fundamental electric charge e and any magnetic charge q must exhibit the relationship $eq = nh/\mu_0$ whence any monopoles in our universe must be quantized in units of $q_D = h/\mu_0 e$. This follows from the monopole being attached to a Dirac string, which has to be unobservable[17]. By contrast, the string soup characteristic of spin ice at low temperature

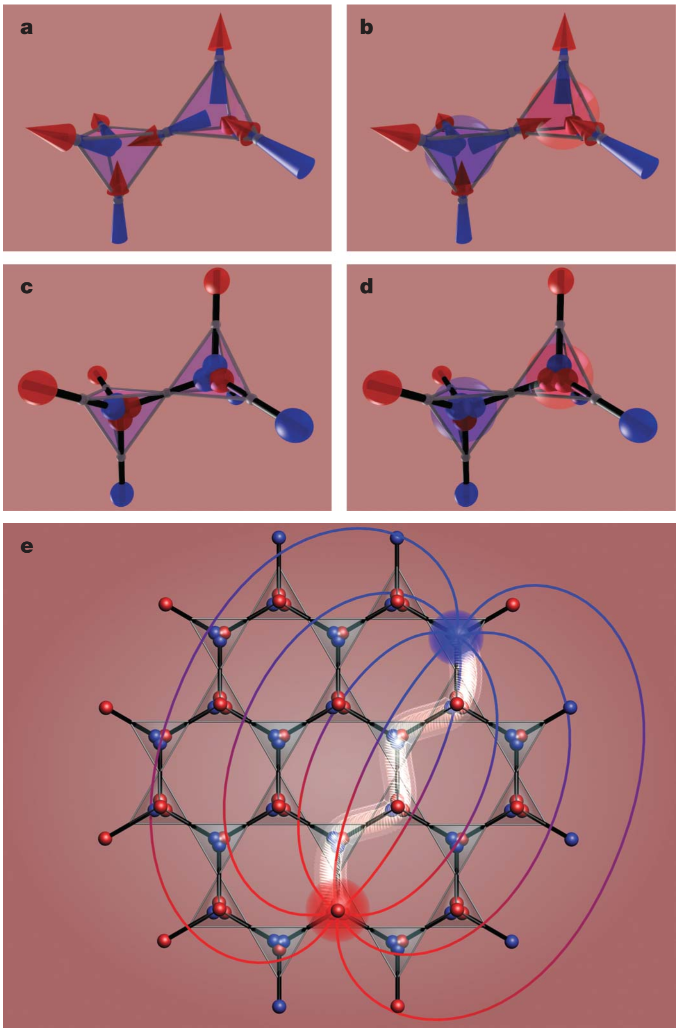

Figure 2 | Mapping from dipoles to dumbbells. The dumbbell picture (**c**, **d**) is obtained by replacing each spin in **a** and **b** by a pair of opposite magnetic charges placed on the adjacent sites of the diamond lattice. In the left panels (**a**, **c**), two neighbouring tetrahedra obey the ice rule, with two spins pointing in and two out, giving zero net charge on each site. In the right panels (**b**, **d**), inverting the shared spin generates a pair of magnetic monopoles (diamond sites with net magnetic charge). This configuration has a higher net magnetic moment and it is favoured by an applied magnetic field oriented upward (corresponding to a [111] direction). **e**, A pair of separated monopoles (large red and blue spheres). A chain of inverted dipoles ('Dirac string') between them is highlighted in white, and the magnetic field lines are sketched.

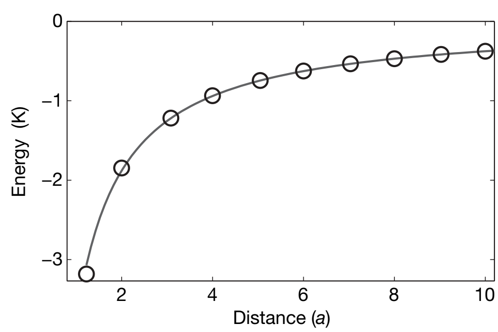

Figure 3 | Monopole interaction. Comparison of the magnetic Coulomb energy $-\mu_0 q_m^2/(4\pi r)$ (equation (2); solid line) with a direct numerical evaluation of the monopole interaction energy in dipolar spin ice (equation (1); open circles), for a given spin-ice configuration (Fig. 2e), as a function of monopole separation.

makes the strings energetically unimportant, although they are observable and are therefore not quantized.

Indeed, the monopoles in spin ice have a magnitude $q_m = 2\mu/a_d = 2(\mu/\mu_B)(\alpha\lambda_C/2\pi a_d)q_D \approx q_D/8{,}000$, where λ_C is the Compton wavelength for an electron, and α is the fine-structure constant. The charge of a monopole in spin ice can even be tuned continuously by applying pressure, because this changes the value of μ/a_d.

The monopoles are sources and sinks of the magnetic field **H**, as is appropriate to the condensed matter setting. More precisely, as in other instances of fractionalization[20], we can define a 'smeared' magnetic charge $\rho_m(\mathbf{R}) = \int d^3\mathbf{r}' \exp\left(-|\mathbf{r}'-\mathbf{R}|^2/\xi^2\right) \nabla\cdot\mathbf{H}$, where $\nabla\cdot\mathbf{H}$ is the divergence of the magnetic field. For a monopole at the origin, separated by $L \gg \xi \gg a$ from any other monopoles, this gives $\rho_m(0) = \pm q_m$. The form of the magnetic induction **B** is also monopolar, but with the important difference that a compensating flux travels along the (unquantized) 'Dirac string' of flipped dipoles created along with the monopole (see Fig. 2).

Our magnetic monopoles would in principle show up in one of the best-known monopole searches, the Stanford experiment to detect fundamental magnetic monopoles from cosmic radiation. This experiment is based on the fact that a long-lived current is induced in a superconducting ring when a monopole passes through it[8]. We can easily check that the presence of the Dirac string of flipped dipoles is immaterial to the establishment of a current.

The above observations are the central qualitative results of our work: ice-rule-violating defects are deconfined monopoles of **H**, they exhibit a genuine magnetic Coulomb interaction (see equation (2)), and they produce Faraday electromotive forces in the same way as elementary monopoles would.

We re-emphasize that the ice rule alone does not permit a consistent treatment of the excited states of the physical problem: crucially, the energetic interaction between our defects is absent altogether. Also, in previous discussions of the purely ice-rule problem and related short-range problems[11–13] it has been noted that the defects do acquire a purely entropic Coulomb (that is, $1/r$) interaction, which has a strength that vanishes proportionally to T at low temperatures. This interaction will be present in addition to the magnetic Coulomb interaction discussed in this paper, and is clearly much smaller as $T \to 0$. Also, it will not be accompanied by a magnetic field, it will not renormalize the monopole charge, and it will not be felt by a stationary magnetic test particle that is embedded in the lattice but is not attached to a lattice site.

The most satisfactory way to demonstrate the presence of a monopole would be to measure the force on magnetic test particles, say by a Rutherford scattering experiment or by clever nanotechnological means. Unfortunately, given the lack of elementary magnetic monopoles, we would have to use dipoles as test particles, which significantly weakens such signatures.

An alternative strategy is to look for consequences of the presence of magnetic monopoles in the collective behaviour of spin ice. This is most elegantly achieved by applying a magnetic field in the [111] crystallographic direction. Such a field acts as a (staggered) chemical potential (see Fig. 2), favouring the creation of monopoles of a given sign on either sublattice of the diamond lattice.

We thus have a tuneable lattice gas of magnetic monopoles on the diamond lattice. The basic structure of the phase diagram as a function of magnetic field and temperature can be inferred from work by Fisher and collaborators[21] in the context of ionic lattice gases and Coulombic criticality. At high T, there is no phase transition but a continuous crossover between the high- and low-density phases as the chemical potential is varied. At low T, a first-order phase transition separates the two regimes. This transition terminates in a critical point at (h_c, T_c), not unlike the liquid–gas transition of water. This serves as a useful diagnostic, because the liquid–gas transition is absent for a nearest-neighbour spin-ice model, in which defects interact only entropically. In that case, it is known that there cannot be a first-order transition in the limit of low T (ref. 22).

To confirm this scenario, we have demonstrated by Monte Carlo simulations that the actual phase diagram of dipolar spin-ice model has precisely this structure. To rule out the appearance of the liquid–gas transition being due to effects introduced by the approximations leading to equation (2), we simulated directly the original dipolar spin-ice model, equation (1). The resulting phase diagram is depicted in Fig. 4. The critical endpoint is located around $(T_c, h_c) = (0.57 \pm 0.06\,\text{K},\ 0.86 \pm 0.03\,\text{T})$. The error bars are mainly due to finite-size effects, as the intensive nature of the simulations of long-range dipolar interactions prohibits simulating very large systems.

This scenario is indeed observed experimentally in spin-ice materials[6,7], and our results provide a natural explanation. Spin ice in a [111] magnetic field is a problem that has already attracted considerable attention. The low-density phase of monopoles is known as kagome ice, a quasi-two-dimensional phase with algebraic correlations and a reduced residual entropy[6,7,23]. The high-density phase is an ordered state with maximal polarization along the field direction. Experimental results on the liquid–gas transition and its endpoint are also displayed in Fig. 4 for comparison. Our numerical results are in good qualitative agreement with both experiment and the analytic calculations of ref. 21. Our value of the critical field agrees with ref. 6 to within a few per cent, which is less than the uncertainty due to demagnetization effects[6,7]. However, the experimental value of T_c is about a third lower than the numerical one, most probably due to farther-neighbour (exchange) interaction terms, which—although small—can shift the location of a transition temperature considerably[10].

The presence of a liquid–gas transition was noted to be very remarkable because there are few, if any, other experimentally known instances in localized spin systems[6]. No mechanism was known to account for this phenomenon, and our theory of magnetic monopoles fills this gap.

The existence of magnetic monopoles in a condensed matter system is exciting in itself. (The monopoles appearing in the interesting work on the anomalous Hall effect are not excitations and do not involve the physical magnetic field[24].) Moreover, these monopoles are a rare instance of high-dimensional fractionalization, of interest

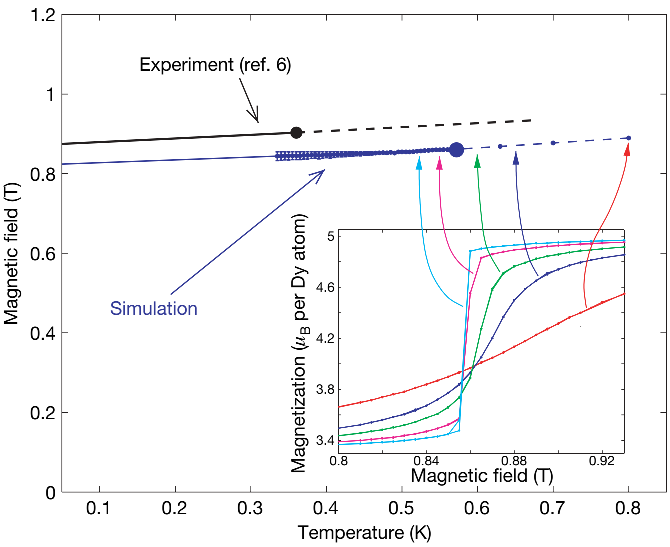

Figure 4 | Phase diagram of spin ice in a [111] field. The location of the monopole liquid–gas transition from numerics (blue line) compared to experiment (black line; ref. 6). The solid lines are first-order transitions terminated by critical endpoints (filled circles). The dashed lines are crossovers. The inset shows magnetization curves showing the onset of first-order behaviour as the temperature is lowered. Our simulations cover the range $0.335\,\text{K} < T < 0.8\,\text{K}$ for 1,024 spins. At the lowest temperatures, the parallel tempering code we use in our simulations of the Ewald-summed dipolar interaction no longer completely suppresses the hysteresis, and we have extended the first-order transition line using Clausius–Clapeyron.

in fields as diverse as correlated electrons and topological quantum computing[25]. We hope our analysis will encourage experiments aimed at directly detecting these monopoles. There are many avenues to explore in search of useful signatures, among them scattering, transport and noise measurements, and flux detection.

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